

Pinned interface dipole-induced tunneling electroresistance in ferroelectric tunnel junctions

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Abstract

Based on the structure predicted in a ferroelectric tunnel junction(FTJ) in the resent density functional theory study[Phys. Rev. B **85** 125407(2012)], we investigate the electron transport through the FTJ with asymmetric interfaces, i.e., one interface dipole is pinned and the other interface dipole is switchable. Tunneling electroresistance(TER) can be induced due to the nonswitchable interface dipole in FTJs with symmetric electrodes. Compared with the dependence relationship between TER and the polarization of switchable interface, TER is not sensitive to the variation of the polarization of pinned interface. A large TER can be achieved when the pinned polarization points to the ferroelectric film and low interface dielectric constants. In addition, effect of electrode on TER in the structure is also discussed.

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I. INTRODUCTION

The existence of ferroelectricity in nanometer-thick films makes it possible to use ferroelectrics as barrier in tunnel junctions[1], FTJ with polarization switching show important electronic transport properties, such as giant TER[2, 3], solid-state memories[4]. Furthermore, switchable polarization of the tunnel barrier allows for controlling of the transport spin polarization if the electrodes are ferromagnetic[5, 6].

In theory, Tsymbal's group proposed that the surface charges in the ferroelectric are not completely screened by the adjacent metals and therefore the depolarizing electric field in the ferroelectric is not zero. If a FTJ is made of asymmetric metal electrodes which have different screening lengths, this lead to the asymmetry in the potential profile for the opposite polarization directions. Thus, the potential seen by transport electrons changes with the polarization reversal which lead to a giant TER effects[1, 7]. In general, the interface is intrinsic and inevitable in the metal-ferroelectric-metal structure during preparing process. The presence of interfaces will reduce the symmetry of a FTJ, and may produce electric dipoles at the interface. Duan[8]studied ultrathin KNbO_3 ferroelectric films placed between metal electrodes, either SrRuO_3 (SRO) or Pt, it is found that the bounding at the ferroelectric-metal interfaces imposes severe constrains on the displacement of atoms, and induces an fixed interface dipole moment, which is electrode dependent and non-switchable. Stengel and Spaldin[9] demonstrated the existence of a dielectric dead layer in SRO/ SrTiO_3 /SRO nanoscale capacitor, which is only one or two atomic monolayers at the interface. Based on first-principles and model calculations, Wang[10] investigated a ferroelectric dead layer near the interfaces that is nonswitchable, and predicted mechanism for the dead layer at the interface controls the critical thickness for ferroelectricity in systems with polar interfaces. Liu[11]found that a BaO/RuO_2 interface termination sequence in SRO/ BaTiO_3 /SRO epitaxial heterostructure grown on SrTiO_3 (STO) can lead to a nonswitchable polarization state for thin BaTiO_3 (BTO) films due to a fixed interface dipole, while the interfacial polarization at the TiO_2/SrO interface is switchable. And such asymmetric interfaces may destroy the stability of one of the polarization states, making the system only monostable in zero applied field and therefore nonferroelectrics. Their first-principles and phenomenological modeling prediction and experimental measurments[12] confirm that switchable ferroelectric polarization can be achieved by inserting a thin STO layer at BaO/RuO_2 interface to eliminate the

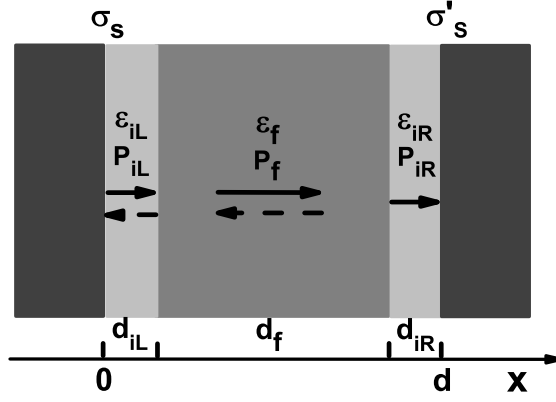


FIG. 1: Sketch of the FTJ with a pinned interfacial dipole P_{iR} , whose direction can be arbitrarily preassigned. P_{iL} and P_f stand for the polarization of the left interface and the medial ferroelectric film, and they can be reversed by an external electric field.

nonswitchable interface dipole. Previous studies are focused on the interfacial effect on the polarization[10] and critical thickness[11] of the ferroelectric barrier. Recently, we have investigated electron transport in the FTJ with nonswitchable interface polarizations at both the left and right metal-ferroelectrics interfaces[13]. However, the transport properties of FTJ with asymmetric interface, i.e., one interface dipole is switchable and another interface dipole is nonswitchable, has not been studied, and such a structure has been predicted by first-principles calculations in epitaxial SRO/BTO/SRO heterostructure[11]. In this paper, effects of electrode, interface polarization and interface dielectric constant on the TER in FTJs with a pinned interface dipole are investigated in detail. We obtain that the pinned interface dipole can induce electroresistance in a FTJ with symmetric electrodes. Furthermore, comparison between switchable and nonswitchable interface dipoles on TER are also given.

II. MODEL AND THEORY

The structure of a FTJ with two interfaces between the metal electrodes and ferroelectric barrier is shown in Fig. 1. It is assumed that the polarizations of the interface and ferroelectric barrier are perpendicular to the electrode, which can be realized by designing the misfit compression stress from the substrate. Similar to the results of first-principles calculations in SRO/BTO/SRO nanostructure, we assume the dipole in the right interface is pinned, and its direction can be arbitrarily selected as pointing to the left or right. Once

selected, its direction cannot be switched under applied electric fields. The dashed arrows in Fig. 1 indicate that the corresponding polarizations can be reversed by an applied field. We do not consider inhomogeneous polarizations throughout the barrier for simplicity, and the interface layer is selected as one unit cell ($d_{iL}=d_{iR}=4\text{\AA}$) [13, 14].

According to the Thomas-Fermi model, the screening potential within the left and right electrodes can be given by

$$\varphi(x) = \begin{cases} -\frac{\sigma_s \lambda_L}{\epsilon_L} e^{x/\lambda_L}, & x \leq 0, \\ \frac{\sigma_s \lambda_R}{\epsilon_R} e^{-(x-d)/\lambda_R}, & x \geq d. \end{cases} \quad (1)$$

Here $\lambda_L(\lambda_R)$ is the Thomas-Fermi screening length of the left(right) electrode, ϵ_L and ϵ_R are the dielectric constants of the left and right electrodes. σ_s stands for the screening charge per unit area in the metal electrode which is the same in the left and right electrodes due to the charge conservation condition. The continuity conditions of electric displacement and electric potential at the boundaries are

$$\begin{cases} \epsilon_{iL} E_{iL} + P_{iL} = \sigma_s, & x = 0, \\ \epsilon_f E_f + P_f - (\epsilon_{iL} E_{iL} + P_{iL}) = 0, & x = d_{iL}, \\ \epsilon_{iR} E_{iR} + P_{iR} - (\epsilon_f E_f + P_f) = 0, & x = d_{iL} + d_f, \\ \epsilon_{iR} E_{iR} + P_{iR} = \sigma_s, & x = d_{iL} + d_f + d_{iR}, \\ E_{iL} d_{iL} + E_f d_f + E_{iR} d_{iR} = \varphi(0) - \varphi(d), \end{cases} \quad (2a)$$

$$(2b)$$

where E_{iL} , E_f and E_{iR} are the depolarization fields in the left interface, the barrier and the right interface, respectively. P_{iL} , P_{iR} and P_f are the polarization of the interfaces and barrier. From Eqs. (1) and (2), the depolarization fields and then the electrostatic profile $\varphi(x)$ ($0 < x < d$) are obtained as follows

$$\begin{aligned} E_{iL}^d &= \frac{\sigma_s - P_{iL}}{\epsilon_{iL}}, \\ E_f^d &= \frac{\sigma_s - P_f}{\epsilon_f}, \\ E_{iR}^d &= \frac{\sigma_s - P_{iR}}{\epsilon_{iR}}, \end{aligned} \quad (3)$$

and

$$\varphi(x) = \begin{cases} \varphi(0) - E_{iL}^d x, & 0 < x \leq d_{iL}, \\ \varphi(d_{iL}) - E_f^d (x - d_{iL}), & d_{iL} < x \leq d_{iL} + d_f, \\ \varphi(d_{iL} + d_f) - E_{iR}^d (x - d_{iL} - d_f), & d_{iL} + d_f < x \leq d, \end{cases} \quad (4)$$

where

$$\sigma_s = \frac{\frac{P_{iL} d_{iL}}{\epsilon_{iL}} + \frac{P_f d_f}{\epsilon_f} + \frac{P_{iR} d_{iR}}{\epsilon_{iR}}}{\frac{\lambda_L}{\epsilon_L} + \frac{d_{iL}}{\epsilon_{iL}} + \frac{d_f}{\epsilon_f} + \frac{d_{iR}}{\epsilon_{iR}} + \frac{\lambda_R}{\epsilon_R}}. \quad (5)$$

The overall potential profile $U(x)$ seen by transport electrons across the junction is the superposition of the electrostatic potential $-e\varphi(x)$, the electronic potential which determines the bottom of the bands in the two electrodes with respect to the Fermi energy E_F , and the potential barrier created by the interfaces and barrier. It is assumed that the interfaces potential and the barrier potential have a rectangular shape of height U_{iL} , U_{iR} and U_f with respect to the E_F [7].

At a small bias voltage the conductance per unit area of the junction can be obtained using Landauer formula[15, 16]

$$G = \frac{2e^2}{h} \int \frac{dk_{\parallel}}{(2\pi)^2} T(E_F, k_{\parallel}), \quad (6)$$

where $T(E_F, k_{\parallel})$ is the transmission coefficient at the Fermi energy for a given transverse wave vector k_{\parallel} , and can be calculated by solving the schrödinger equation for an electron tunneling through the potential barrier $U(x)$ within the formalism of transfer matrix [17, 18]. It is necessary to note again that, as show in Fig. 1, the dipole P_{iL} within the left interface and the spontaneous polarization P_f in the ferroelectric barrier are always in the same direction, and their direction can be reversed under the action of an external electric field, while the dipole P_{iR} in the right interface is nonswitchable. In this paper, the right direction is defined as the positive polarization direction, and the TER ratio is defined as $TER = \frac{G_R - G_L}{G_L}$, where G_L and G_R are conductances of a FTJ for polarization in the barrier pointing left and right, respectively. It is well known that the larger the value TER is, the better the performance of FTJs.

III. RESULTS AND DISCUSSIONS

Firstly, the TER of a FTJ with a pinned interface dipole and symmetric electrodes is numerically calculated. Pt and SRO electrodes are used in our study to simulate effect of electrodes on TER, as well as the effect of nonswitchable interface dipole on TER. The screening length of Pt(SRO) is selected as $0.4\text{\AA}(0.75\text{\AA})$, and the dielectric constants of Pt and SRO electrodes takes the values ϵ_0 and $8.85\epsilon_0$, respectively[18]. The Fermi energy is chosen as $E_F = 3.0eV$, and the barrier heights for the interface and FE barrier are taken as $U_{iL} = U_{iR} = 0.6eV$ and $U_f = 0.6eV$ [13]. The polarization, the dielectric constant and the thickness of the ferroelectric film are chosen as $P_f = 0.26C/m^2$, $\epsilon_f = 90\epsilon_0$ and $d_f = 2nm$ in

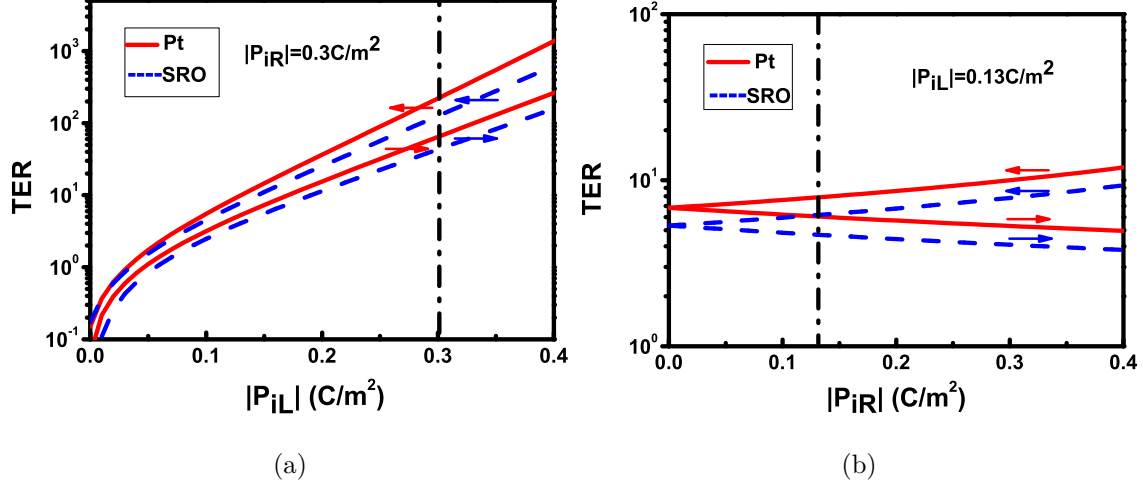


FIG. 2: (Color online) TER as a function of the magnitude of (a) P_{iL} and (b) P_{iR} . Solid lines denote the TER for Pt electrodes, dashed lines for SRO electrodes, and dash-dot line is drawn to indicate where the magnitude of the two interfaces are equal. Arrows represent the direction of nonswitchable P_{iR} , which is assumed to preassign only one of the two directions.

this paper.

In Fig. 2, TER as a function of the polarizations of switchable interface dipole P_{iL} and pinned interface dipole P_{iR} are plotted. The arrows near the lines stand for the direction of P_{iR} , which is preassigned. The vertical dash-dot line in Fig. 2 denotes where the magnitude of polarization of the left interface and the right interface are equal, however, the left interface dipole is switchable, and the right one is nonswitchable. If the two interfaces have the same polarization and both of them are switchable, then TER of the corresponding FTJ will be zero[7]. From the intersection of dash-dot line and TER curves in Fig. 2, one can see that TER is not zero. Therefore, TER can be induced by asymmetric interface dipoles, such as the asymmetry in switching. Moreover, TER always increases with the increase of the magnitude of switchable dipole P_{iL} , while TER increases with increase of P_{iR} only when the nonswitchable dipole P_{iR} pointing to the barrier(left), and TER decreases as P_{iR} increases when P_{iR} points to the electrode(right). Compared Fig. 2(a) with 2(b), we find that TER is more sensitive to the change of P_{iL} than the change of P_{iR} , it is because that the left interface dipole P_{iL} is switchable and has the same direction with P_f , therefore the increase of P_{iL} is equal to enhance the effective spontaneous polarization of the FTJ, then, results in an increase of TER. Under the same conditions, the nonswitchable dipole P_{iR} pointing to barrier favors a large TER, which is consistent with our previous results[13]. As the effect

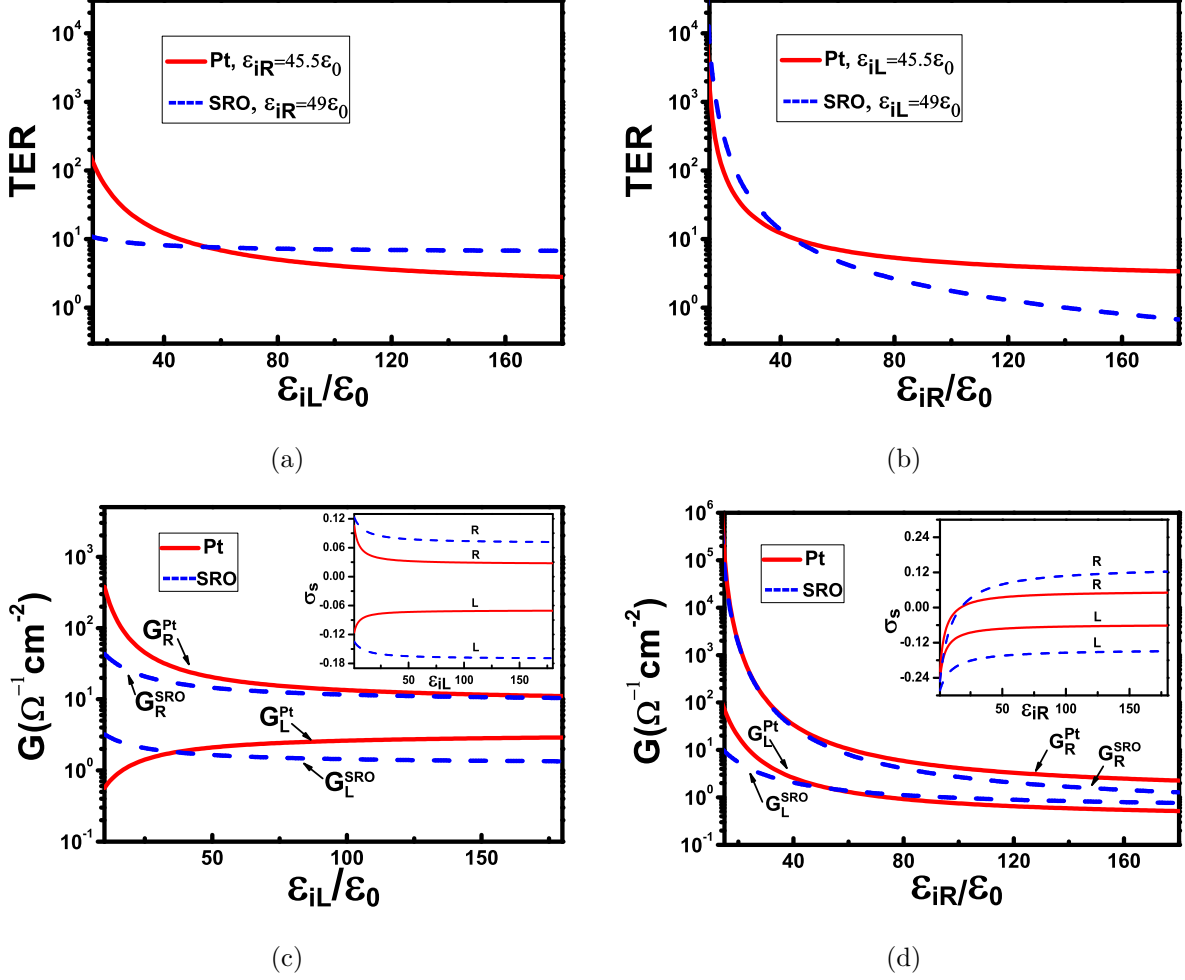


FIG. 3: (Color online) TER as a function of (a) the dielectric constant of the left interface ϵ_{iL} and (b) the dielectric constant of the right interface ϵ_{iR} , and tunneling conductance as a function of (c) ϵ_{iL} and (d) ϵ_{iR} . Solid lines denote the case for Pt electrodes and dash lines for SRO electrodes. The inset in (c) and (d) represent the change of σ_S with ϵ_{iL} and ϵ_{iR} respectively. The letter L(R) in the insets stand for P_f pointing to left(right). Here, the interface polarization are taken as $|P_{iL}| = 0.13C/m^2$ and $P_{iR} = -0.3C/m^2$.

of electrode is concerned, the FTJ with Pt electrodes will have a large TER than that with SRO electrodes when the interfacial dielectric constants take the average value between the dielectric constant of electrode and that of ferroelectric film. Here, $\epsilon_{iL} = \epsilon_{iR} = 45.5\epsilon_0$ for Pt electrodes and $\epsilon_{iL} = \epsilon_{iR} = 49\epsilon_0$ for SRO electrodes in Fig. 2.

To investigate the dependence of TER on the interfacial dielectric constants, polarization of the left and right interfaces are fixed as $|P_{iL}| = 0.13C/m^2$ and $P_{iR} = -0.3C/m^2$ respectively. The minus sign of P_{iR} means that the pinned dipole P_{iR} is assumed to point

to left. The above interface polarization are carefully chosen based on the first-principles calculations for the SRO/BTO/SRO junction[11].

It is shown, from Fig. 3(a) and (b), that TER decreases with the increasing of interfacial dielectric constants whether the corresponding interface dipole is reversal or not. Furthermore, TER varies remarkably within the range of low ϵ_{iL} and ϵ_{iR} , however, within the range of large interfacial dielectric constant, TER changes slowly. Compared Fig. 3(a) with 3(b), we can obtain TER increases about three orders as ϵ_{iR} decreases, while TER is only raised one order as ϵ_{iL} decreases. We also calculate TER for small magnitude of P_{iR} , and TER will increase slowly with the decrease of ϵ_{iR} . Therefore, the remarkable increase of TER when ϵ_{iR} decreases in Fig. 3(b) is due to the large quotient $\frac{P_{iR}d_{iR}}{\epsilon_{iR}}$ in Eq. 5, i.e., the great change of σ_s . From Fig. 3(a), one can also find that, compared with SRO electrodes, TER is more sensitive to the variation of ϵ_{iL} for Pt electrodes(solid line). On the contrary, in Fig. 3(b), TER is more sensitive to the change of ϵ_{iR} for SRO electrodes(dashed line). The above phenomena are caused by the variation of the distribution of screening charges σ_s , which is shown in the insets of Fig. 3(c) and (d). In the inset of Fig. 3(c), σ_s decreases when ϵ_{iL} decreases for Pt electrodes when the barrier polarization points to left, and the decrease of σ_s results in the decrease of G_L^{Pt} and then the steep increase of TER for a FTJ with Pt electrodes within low ϵ_{iL} range. With the decreasing of ϵ_{iR} in the inset of Fig. 3(d), σ_s decreases more quickly for SRO electrodes when the barrier polarization points to right in comparison with other cases, which brings a steep increase of G_R^{SRO} and TER in Fig. 3(d) and in Fig. 3(b), respectively. As effect of electrodes on TER is concerned, SRO electrode is preferable to Pt electrode within the range of large ϵ_{iL} in Fig. 3(a), and for low ϵ_{iL} , Pt electrode is preferable. Thing will be completely opposite for the choice of electrode within the large and low scopes of ϵ_{iR} in Fig. 3(b).

In summary, the ferroelectric tunneling junction with symmetric electrodes is studied by using of Thomas-Fermi model and quantum tunneling theory, and it is found that TER is induced in the above FTJ with a pinned interface dipole. TER as a function of the interface polarization and dielectric constant is given, we obtain that TER can be great improved by enhancing the polarization of the switchable interface dipole, while TER is not sensitive to the change of polarization of the nonswitchable interface dipole. A large TER is found when the nonswitchable interface dipole points to the ferroelectric barrier. A significant TER can be obtained for low interface dielectric constants. The electrode effect on TER is

also presented. In one word, the model of a FTJ with only one pinned interface dipole is a practical model, and effects of the switchable interface and the nonswitchable interface on TER are different. We hope our studies can provide suggestion and guide on the preparation of high performance FTJs.

Acknowledgments

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Figures' Caption

FIG. 1: Sketch of the FTJ with a pinned interfacial dipole P_{iR} , whose direction can be arbitrarily preassigned. P_{iL} and P_f stand for the polarization of the left interface and the medial ferroelectric film, and they can be reversed by an external electric field.

FIG.2: (Color online) TER as a function of the magnitude of (a) P_{iL} and (b) P_{iR} . Solid lines denote the TER for Pt electrodes, dashed lines for SRO electrodes, and dash-dot line is drawn to indicate where the magnitude of the two interfaces are equal. Arrows represent the direction of nonswitchable P_{iR} , which is assumed to preassign only one of the two directions.

FIG.3: (Color online) TER as a function of (a) the dielectric constant of the left interface ϵ_{iL} and (b) the dielectric constant of the right interface ϵ_{iR} , and tunneling conductance as a function of (c) ϵ_{iL} and (d) ϵ_{iR} . Solid lines denote the case for Pt electrodes and dash lines for SRO electrodes. The inset in (c) and (d) represent the change of σ_S with ϵ_{iL} and ϵ_{iR} respectively. The letter L(R) in the insets stand for P_f pointing to left(right). Here, the interface polarization are taken as $|P_{iL}| = 0.13C/m^2$ and $P_{iR} = -0.3C/m^2$.